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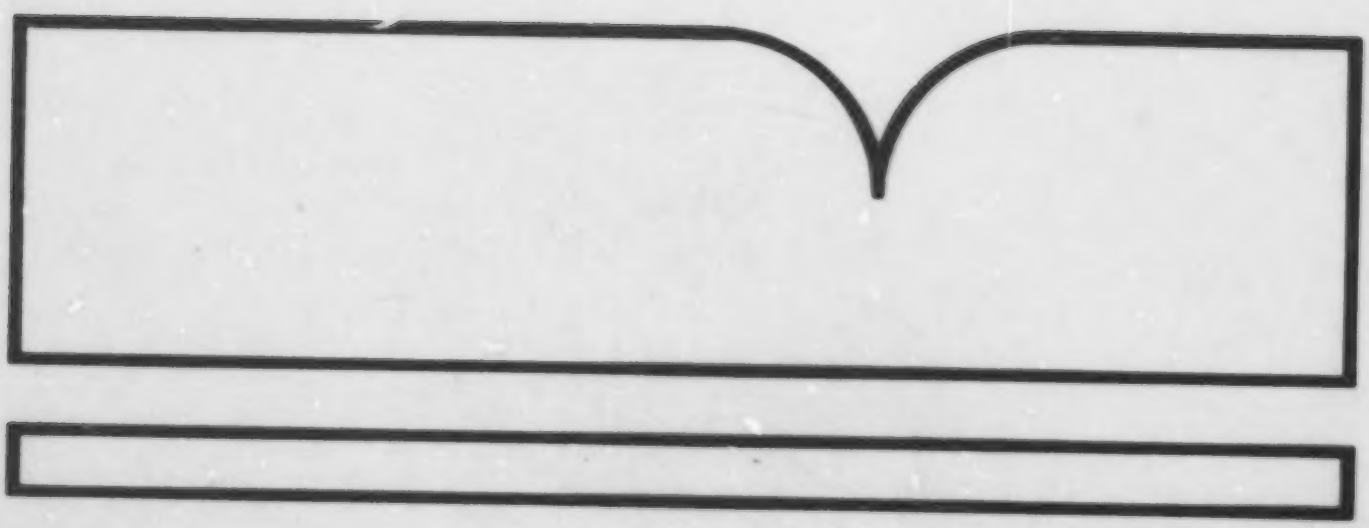

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16. ABSTRACT

In the concept of an "urban plume," the combined air pollutants from several major emission sources in an urban area are considered to be carried, with air masses moving across the city, out over nonurban land or water. A plume may be distinguished from the unpolluted nonurban atmosphere, which is not directly downwind of a city, by its chemical composition and high concentrations of certain pollutants. However, in the eastern U.S., where the most abundant particulate air pollutants in the plume may be sulfate aerosol formed by the oxidation of gaseous sulfur oxides, the urban source areas are spaced closely enough so that the intervening nonurban regions are often under the combined influence of several urban plumes. The resulting generally high nonurban average pollutant concentrations, especially for sulfate, greatly complicate the task of tracing the transport of individual urban plumes, and, of particular importance, of determining the extent of sulfate formation within them.

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SULFATE FORMATION IN URBAN PLUMES*

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INTRODUCTION

In the concept of an "urban plume," the combined air pollutants from several major emission sources in an urban area are considered to be carried, with air masses moving across the city, out over nonurban land or water. A plume may be distinguished from the unpolluted nonurban atmosphere, which is not directly downwind of a city, by its chemical composition and high concentrations of certain pollutants. However, in the eastern U.S., where the most abundant particulate air pollutants in the plume may be sulfate aerosol formed by the oxidation of gaseous sulfur oxides, the urban source areas are spaced closely enough so that the intervening nonurban regions are often under the combined influence of several urban plumes. The resulting generally high nonurban average pollutant concentrations, especially for sulfate, greatly complicate the task of tracing the transport of individual urban plumes and, of particular importance, of determining the extent of sulfate formation within them. Consequently, larger scale field studies may be desirable, such as by networks of nonurban sampling stations throughout the eastern U.S. region, measurements over the ocean downwind of the entire urbanized northeastern U.S., or a comparison of sulfate concentrations in nonurban northern and southern hemisphere locations, in order to determine the magnitude of sulfate formation and the presence of other air pollutants in composite urban plumes. In this paper, the results of such studies carried out at Florida State University will be briefly summarized. A more complete bibliography of pertinent literature is given in the references cited.

THE EASTERN UNITED STATES NONURBAN SULFATE AEROSOL

A starting point for tracing the formation of sulfate in urban plumes and its transport over nonurban regions is to document the concentration of nonurban sulfate in the eastern U.S., its variability in space and time, the relation between sulfate concentration and other observable quantities such as visibility, and evidence for the dispersion of the combined nonurban sulfate-laden air mass outward in its fringe areas, such as into Florida. Two examples of such investigations are summarized here.

1. Cohn[1] has reported the measurement of aerosol sulfur concentrations from a network of sampling stations ranging from Kansas to New Hampshire, with most

*Portions of the research reviewed here have been supported by the U.S. Environmental Protection Agency, the Florida Sulfur Oxides Study, the National Institutes of Health, the National Science Foundation, the North Atlantic Treaty Organization, the U.S. Office of Naval Research, and the U.S. Department of Energy.

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stations located between St. Louis, Missouri, and Pennsylvania. The concentrations, which were measured in samples collected by the time sequence filter "streaker" sampler using proton-induced x-ray emission (PIXE), showed evidence of large-scale coherence between sites. Such coherence is expected if sulfur oxides and sulfate particulate matter are transported efficiently on a regional scale. The concentrations at the different sampling stations may be plotted on maps at two-hour intervals, the time resolution offered by the streaker samplers, and, considering the air flow regimes inferred from published data of the National Weather Service, the concentrations exhibit patterns that can be contoured. The contoured patterns are seen to vary smoothly with time, suggesting that large-scale transport of aerosol and gaseous sulfur oxides is taking place between urban centers in response to air flow.

Such patterns provide evidence that significant reductions in atmospheric visibility occur when aerosol sulfur concentrations rise above about 2 $\mu g/m^3$ (equivalent to 6 $\mu g/m^3$ sulfate, $SO_4^=$). In order to examine this possibility quantitatively, Cohn selected measurements made on two successive days, July 19 and 20, 1976, at the seven locations listed in TABLE 1. On these days, a strong west to east component of air flow was present without rainfall. Early on July 19, a frontal system was situated to the south of the sampling network and was moving toward the southeast, and late on July 20, another frontal system was approaching from the northwest. However, no frontal activity occurred within the sampling network during these days. Relative humidity was between 70% and 90% at all seven stations on the mornings of both days.

From TABLE 1, it is seen that visibility on July 19 ranged from 6 to 15 miles but on July 20 ranged between 2 and 6 miles, except for the westernmost site in Kansas. A more complete study of all visibility data throughout this region shows that on the second day, visibility was generally poor throughout the region, with frequent reports of haze, in contrast to the first day, when only at selected locations were poor visibility and haze reported. No systematic difference between relative humidity on the two days is apparent. The aerosol sulfur concentrations summarized in TABLE 1 indicate that at Manhattan, Kansas, the concentrations were much lower than at other sites, but from St. Louis eastward, the concentrations were generally a few thousand nanograms per cubic meter. Especially in the easternmost parts of the network, the concentrations were substantially higher on the second day. A more thorough analysis

TABLE 1

VISIBILITY, RELATIVE HUMIDITY, AND AEROSOL SULFUR CONCENTRATIONS AT NETWORK SAMPLING SITES, 0600 HOURS, JULY 19 AND 20, 1976

Station Number	Location	Visibility (miles)		% Relative Humidity		Sulfur Concentration (ng/m^3)	
		7/19	7/20	7/19	7/20	7/19	7/20
1	Manhattan, Kansas	15	15	90	90	410	600
2	St. Louis, Missouri	6	6	71	76	3900	2100
4	Argonne, Illinois	9	6	73	74	2500	5700
5	Remington, Indiana	10	4	76	71	—	5700
7	Angola, Indiana	8	6	90	84	—	3300
9	Delaware, Ohio	15	2	81	90	700	2300
10	Meadville, Pennsylvania	15	3	70	84	1300	2700

of the complete time record, with 2-hour resolution, of sulfur concentrations on both days at all sites and the corresponding variations in visibility, together with the results presented in TABLE 1, indicates that aerosol sulfur concentration is a significant factor in reducing atmospheric visibility. The analysis permits a provisional estimate to be made that when sulfur exceeds about 2000 ng/m^3, and the relative humidity is sufficiently high, a significant reduction in visibility occurs. High relative humidity with low sulfur concentration is not associated as strongly with visibility reduction, nor is high sulfur concentration in very dry air. The facts that, on July 20, poor visibility and haze were reported over the very large area of the network and that the correspondences in sulfur concentrations between sites suggested long range transport indicate that the sulfate formed in urban plumes from different urban areas may combine and contribute to a general visibility reduction over the region under appropriate humidity conditions.

2. A sampling network in the state of Florida offers an excellent opportunity to watch for signs that the composite eastern U.S. sulfate aerosol mass can migrate into Florida from the north. In a sampling network[2] illustrated in FIGURE 1, measurements of the aerosol sulfur concentration as a function of particle size were made over three selected two-week intervals in 1976 and 1977.[3] The average concentrations found for the first period, July–August 1976, for each of the particle size fractions are shown in

FIGURE 1. Index map of Florida showing locations of ground-level sampling sites. Samplers at PN, GV, JAX, KS, AR, PH, and MB were located at small airports, and at TL, TPA, and MIA within the cities.

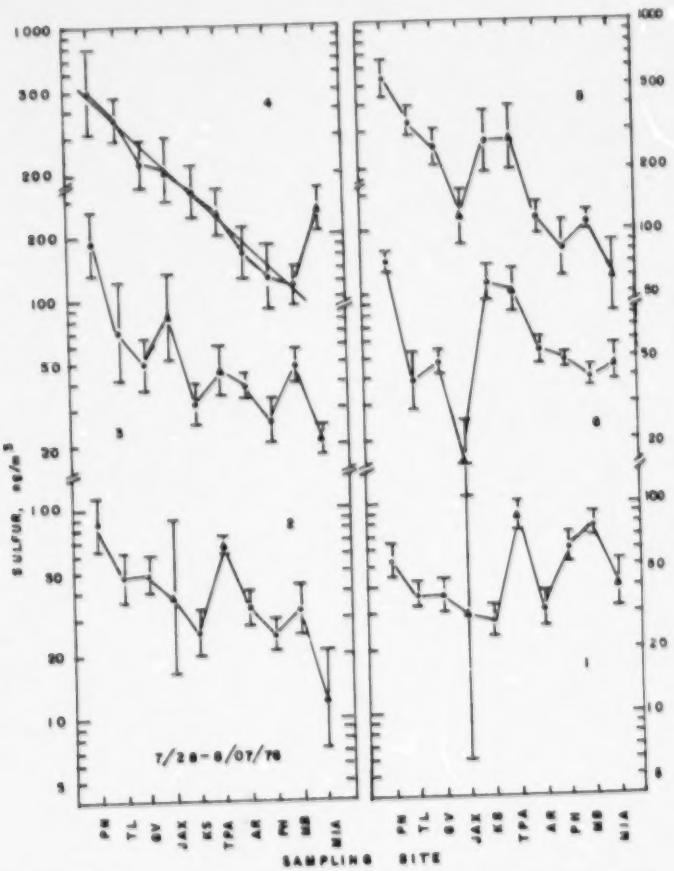


FIGURE 2. Sulfur during the summer, July–August 1976, period in each particle size range 6, < 0.25 μm; 5, 0.25–0.5 μm; 4, 0.5–1 μm; 3, 1–2 μm; 2, 2–4 μm; 1, > 4 μm aerodynamic diameter. Points represent geometric means, with the standard deviations of the means, for groups of approximately six samples taken. Points are plotted against sampling site index. Absolute calibration of concentrations is significant to a factor of 2.

FIGURE 2. In this plot, the average sulfur concentration is found to exhibit a trend with sampling location, from Pensacola to Miami along the length of Florida. If a sampling site index is used as the horizontal coordinate, the concentration in size class 4, 0.5–1 μm aerodynamic diameter, is found to exhibit a monotonic decrease from northwest to south Florida. No marked difference between urban and nonurban sites is seen in the trend for this particle size range. Less smooth trends are seen for some of the other size classes as well, although for the largest- and smallest-particle size classes, a monotonic decrease from northwest to south is observed. Sea salt sulfate, which may contribute much of the largest particle sulfate, comprises a minor amount of the total

aerosol sulfur concentration observed at any of the sites. The fine-particle size range, which comprises the bulk of the sulfur concentration observed at any location, may be formed by gaseous sulfur oxide conversion to sulfate in the atmosphere, ultimately traceable to terrestrial pollution sources.

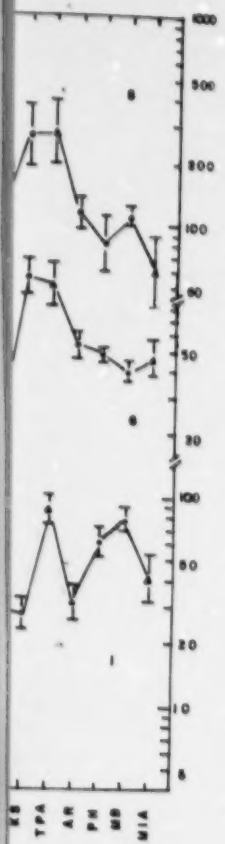
A careful study of meteorological conditions at each location indicates that air masses from states lying to the north of Florida pass over the north Florida sites more frequently than over those sites in the southern part of the state. Thus, in the averaging of approximately six samples collected at each site over the two-week period, a greater percentage of samples of northern air is represented in northern Florida than in the south. Thus, an apparent decreasing average influence of north air flow regime is seen as one moves southward in the state. The decreasing sulfur concentration suggests that Florida is bathed chronically in a sulfate aerosol concentration gradient, due to advection of sulfate formed in combined urban plumes of cities in the eastern U.S. Superimposed on this regional scale gradient may be local urban effects within Florida. However, apparently in the 0.5–1 μm diameter range, these influences are relatively minor, although in some of the other size classes they may be dominant.

Measurements in December 1976 and April 1977 tend to reaffirm the general interpretation presented here. Cascade impactor samples from December show a less pronounced, though still significant, sulfur concentration decrease from north to south of a magnitude that is expected during this season of deeply penetrating cold fronts. Streaker samples collected during the December and April periods allow a detailed examination of the correspondence of sulfur concentration at specific times with air flow patterns inferred from synoptic weather charts.[4] The times when highest sulfur concentrations are observed are generally those when northerly air flow trajectories are likely. In addition, some occurrences of high sulfur concentrations in Tampa are also apparently due to local air pollution effects, although such local effects have not yet been observed at the other Florida locations. The general finding that Florida's nonurban sulfate background is due primarily to transport of sulfate formed in urban plumes from outside the state to the north is of considerable importance in designing strategy for sulfate air pollution control. The existence of a gradient from north to south in sulfate concentration also suggests that epidemiological studies may be carried out in Florida in the search for a correlation between air quality and respiratory disease in the population that resides in this gradient.

The Distance Scale of Sulfate Transport

The preceding examples suggest a large distance scale for the formation and transport of sulfate in urban plumes. They also suggest that, on a continental scale, North America may be more or less contaminated with sulfate formed from sulfur oxide emissions in our urban areas. Thus, in order to find examples of "clean air," one might have to travel away from the continent. The study of Florida indicates that the southernmost locations, which are heavily bathed in marine air, have fine particle sulfur concentrations that may be closer to those of the natural atmosphere than can be found elsewhere in the eastern U.S. However, the full extent of urban sulfate transport may not be fully appreciated unless measurements in more remote maritime and continental atmospheres are made. Two examples may illustrate this point.

1. On two occasions, we have had the opportunity to collect aerosol samples using cascade impactors from towers located on oceanic islands.[5,6] Some results from these



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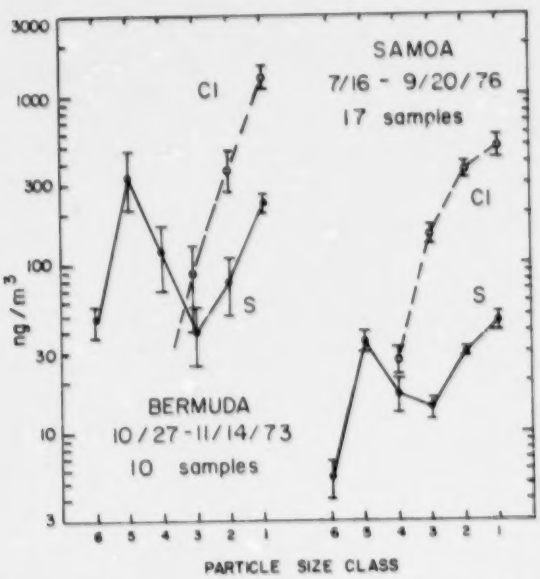


FIGURE 3. Comparison of sulfur and chlorine concentrations, as geometric means ($\pm\sigma_g$), in marine aerosols as a function of particle size, from towers on Bermuda and Samoa. Classes correspond to aerodynamic diameter ranges as in FIGURE 2. (On Samoa, the measured size class 1 concentrations are less than the true values, because of inefficient particle sampling by cascade impactors at high wind speeds.)

studies are shown in FIGURE 3. Bermuda is located in the western North Atlantic Ocean and Samoa is situated in the central South Pacific. In both locations, it may be presumed that the chloride concentration is predominantly due to sea salt injected into the atmosphere by the action of bubbles bursting at the sea surface. At the two locations, the Cl concentrations are quite similar (except for somewhat lower apparent largest-particle Cl concentrations in Samoa, an artifact due to reduced sampling efficiency at high wind speed). The sulfur concentration may be presumed to result from two sources; one, the sea surface, and the other, the conversion from gaseous to particulate sulfate aerosol in the atmosphere. At Bermuda, the concentrations of sulfur are considerably higher than those observed in Samoa, for small particles by about a factor of ten. These results suggest that a composite urban plume of continental dimensions, possibly largely from North America, has transported gaseous sulfur oxides and sulfate aerosol through the atmosphere over the ocean more than one thousand kilometers from the nearest sources. Sulfate formed in the urban plume may be manifested as fine aerosol particles and also possibly as larger particles, if gaseous absorption by marine aerosol droplets is followed by oxidation to sulfate.

Additional measurements made in remote continental atmospheres from South America[7] and southern Africa[8] indicate similar, possibly even lower, aerosol sulfur concentrations than in marine air. These concentrations, 50–100 ng/m^3, are one hundred times lower than concentrations of particulate sulfur often encountered in the

nonurban eastern U.S. atmosphere. These remote atmospheric studies, then, enable us to appreciate how large is the extent of our modification of the composition of the atmosphere in urbanized regions—sulfate concentrations may be one hundred times above the natural level.

2. On a ship midway between Bermuda and New York in the western North Atlantic, Darzi[9] obtained a series of 27 cascade impactor aerosol samples, of 9- to 68-hour sampling times, over a three-week period when air flow varied in its direction, including five samples that were collected approximately downwind of northeast U.S. urban centers. Thus, samples were obtained of a composite urban plume as well as of the atmosphere more typical of the western North Atlantic outside of the urban plume. As with the results in Bermuda, most of the sulfur in the samples was found to be in the fine particle mode, not the coarse particles typical of sea spray, and apparently mostly of pollution origin. However, the five "downwind" samples, collected when a significant fraction of the air had passed over eastern U.S. cities 1–2 days previously, exhibited an anomalously high percentage of the smaller-particle size sulfur contained in the ultrafine < 0.25 μm aerodynamic diameter size range. As shown in FIGURE 4, 50–100% additional ultrafine sulfur, over and above that in the 0.25–2 μm diameter fine range, was observed, whereas the other samples typically had

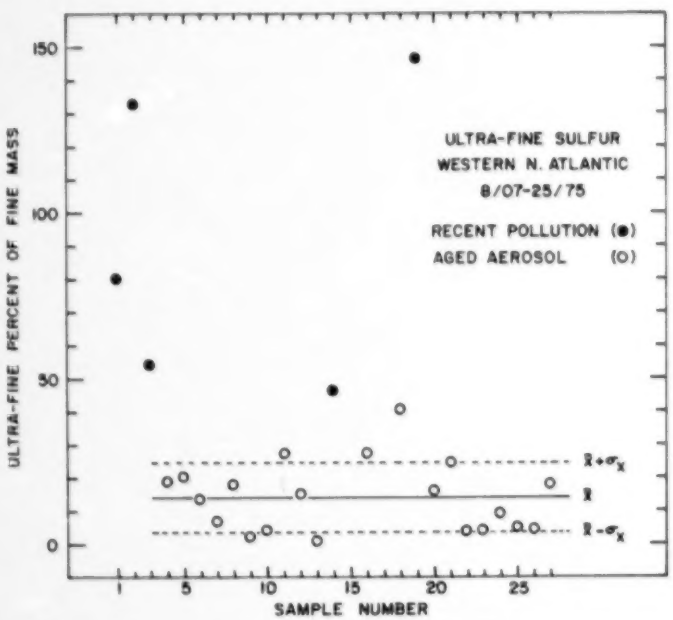


FIGURE 4. Proportions of sulfur in the ultrafine-particle size fraction (class 6, < 0.25 μm) relative to the fine range (classes 5, 4, & 3; 0.25–2 μm) from shipboard on the North Atlantic midway between New York and Bermuda. "Recent pollution" refers to samples collected during times when air trajectories had passed over the New York area 1–2 days previously. "Aged aerosol" refers to samples collected from air that was located over water at least 2 days.

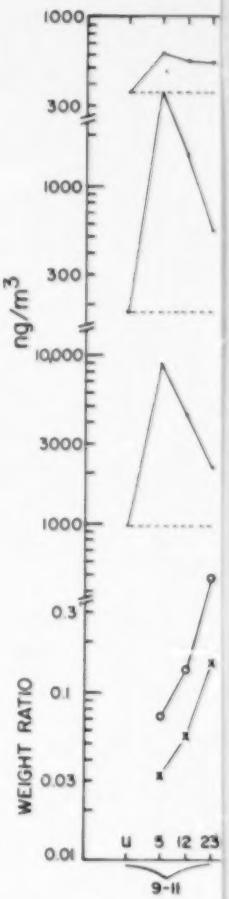
less than 25% additional ultrafine sulfur. These results suggest that either sulfate was being formed directly in the urban plume over the ocean by gas-to-particle conversion, initially producing very small particles, or, if the smallest particles were formed in the urban areas themselves, they had not had sufficient time for coagulation to larger particles during the 1–2 days transport time over the ocean surface. This result of the urban plume could not be detected by comparing total sulfate aerosol concentrations, or even those in the fine-particle size range, since the concentrations were not greater in the "downwind" samples than in the others. Instead, the effect of the plume could be detected by examining the relative percentages of ultrafine-particle size sulfur. Apparently, significant kinetic processes were occurring on a time scale comparable to the transport time over a 1000 km or greater distance out to sea.

The Time Scale of Sulfate Formation in Plumes

The previous examples suggest that the time scale of sulfate formation in plumes may be sufficiently long for transport to occur over hundreds and perhaps thousands of kilometers during the formation process. It would be desirable to design field experiments that would more precisely indicate the time scale and how it varies with geographic and meteorological conditions. Two examples of such field experiments are summarized here.

1. In northwest New Mexico, Ferek[10] directly sampled the plume from a large coal-fired power plant, using ground-based, time sequence, streaker filter samplers. Although the exact direction of air flow was variable, there were times that could later be identified when air flow was consistently along a specified direction. Streaker samplers, which have a two-hour time resolution, were set up in a small array and allowed to operate automatically for several consecutive one-week periods. Then, after examining meteorological records for those time periods when air flow was found to occur in a desired trajectory across the array of samplers, the samples corresponding to a single plume as it passed from one sampler to the next were selected for analysis. In his test of this idea, Ferek examined trends in aerosol sulfur and associated fly ash element concentrations in the plume for successive two-hour periods on August 20, 1976. The results are summarized in Figure 5.

Using an upwind station as a regional background reference, the concentrations of all elements are found to vary with distance downwind at the three sampling sites situated at 5, 12, and 23 km from the power plant stack. Al and Si are representative of several elements found within fine fly ash particles. S represents sulfate formed in the plume from gaseous SO_2, which comprised the bulk of the plume sulfur. By assuming that all constituents are diluted equally as the plume expands during its transport, the relative concentration of aerosol sulfur, aluminum and silicon may represent sulfate formation in the plume. With knowledge of the speed of the plume and the initial stack gas composition of the power plant, the rate of conversion of SO_2 to sulfate can be estimated; for this day, a rate of 0.7% per hour is obtained. Although we may not assume that this value from New Mexico can be considered typical of all conversion rates in all plumes, it is consistent with measurements reported elsewhere in the literature. Moreover, the technique of using ground-based samplers is simple and, in principle, could be applied to other locations. Its further application to plume



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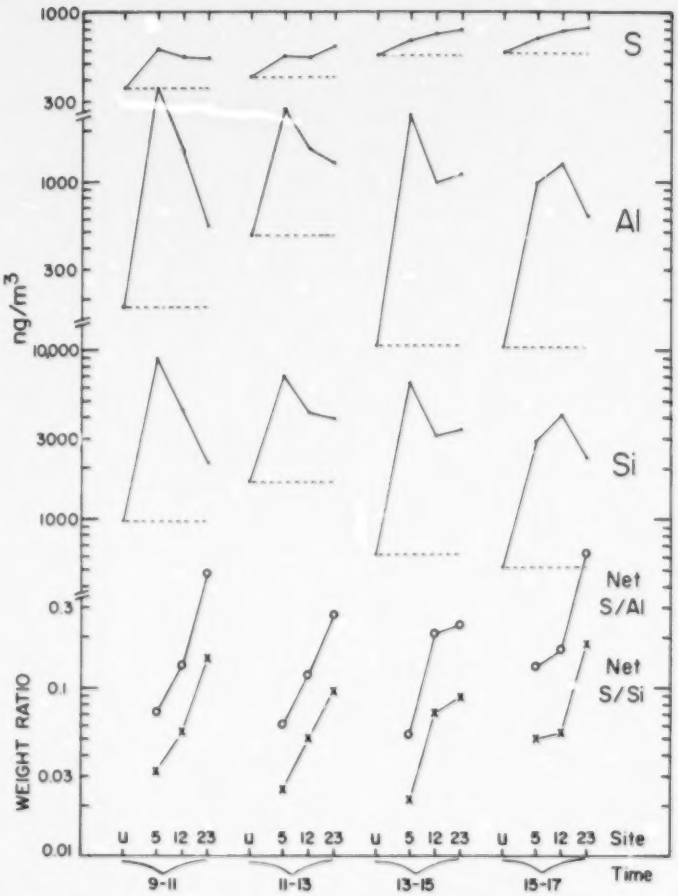

FIGURE 5. Sulfur, aluminum, and silicon concentrations in aerosols from NW New Mexico at ground-level sites located upwind (u) and downwind (at 5, 12, and 23 km) of a large coal-fired electric power plant, during four successive 2-hour intervals on August 20, 1976. Net Al, Si, and S components above upwind concentrations represent fly ash and conversion of gaseous SO_2 to sulfate in the plume. From their ratios, a conversion rate of 0.7% per hour is estimated.

studies may indicate more fully how the rate of sulfate formation in such plumes varies with environmental conditions.

2. The time variability of sulfur concentration in aerosols near the ground in the eastern U.S. is apparently a complicated function of atmospheric environmental factors. In order to examine the atmospheric chemical relationships in detail, we are currently carrying out routine sampling of aerosols at the nine Class I sites of the

EPRI-SURE network ranging from Tennessee to Massachusetts. The samples, collected with streaker samplers, permit PIXE analysis for sulfur and associated trace elements, with 2-hour time resolution.

In FIGURE 6, data from the first two weeks of October 1977 at SURE Site 6, Giles County, Tennessee, are presented.[11] In this example, in many ways typical of nonurban sulfur concentration measurements we have made at other locations in the network, sulfur varies in a seemingly complicated way over a factor of ten in concentration and rarely shows correlation with any of the other trace elements also measured. However, by close examination of the data, several correlations with meteorological data may be discovered. In the first place, the highest concentrations

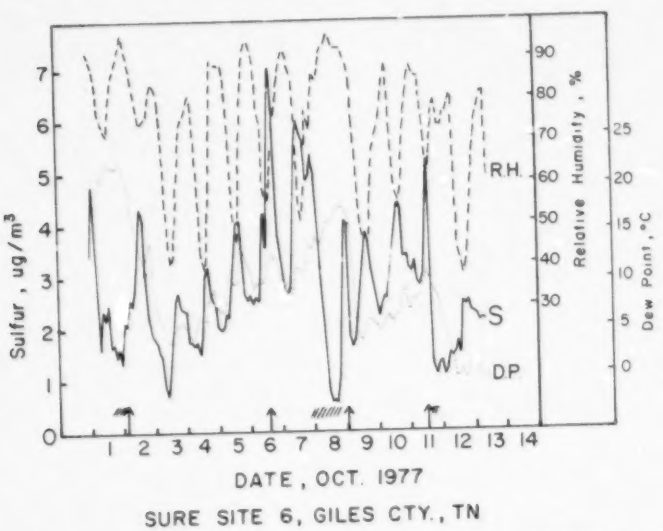

FIGURE 6. Sulfur concentrations measured at 2-hour intervals at SURE Site 6, Giles County, Tennessee, October 1–13, 1977. Also shown are trends of relative humidity, dew point, and times of four frontal passages (arrows) and rainfall (hatched). Sulfur exhibits daytime maxima, when RH exhibits minima and when dew point rises. Sulfur maxima also occur during three of the frontal passages.

often are found near the times of frontal passage. The occurrence of rainfall may tend to decrease concentrations. In this example, additional maxima in sulfur concentrations are seen to occur during the daytime, when relative humidity values exhibit minima. Moreover, the dew point, which is a measure of the absolute humidity of the atmosphere, exhibits maxima at times corresponding to decreasing aerosol sulfur concentration.

At the present time, we cannot present a detailed interpretation of the chemical and dynamic factors that influence sulfate aerosol concentrations in the atmosphere. Undoubtedly, the factors are more than a few in number and may interact in a complex and subtle fashion. In seeking to identify them, it is important to make

aerosol concentration measurements on a time scale comparable to significant variations in humidity, trace gas concentrations, and other atmospheric parameters, which may be such controlling factors. Although at this time we cannot make a decision as to the kinetic mechanisms controlling sulfate concentration, the evidence appears to indicate that mechanisms on both a relatively short time scale, e.g., hourly, and a much longer time scale, e.g., a day or more, may be operative.

Summary

The detection of sulfate formed in urban plumes of cities in the eastern U.S. is complicated by the presence of a high nonurban sulfate background caused by the combined effect of all anthropogenic sulfur oxide sources in the region. This pollution background tends to obscure the contributions of individual urban plumes.

The time period for appreciable SO_2 conversion to sulfates may be comparable to that required for plume transport over tens to hundreds of kilometers. Nonurban submicrometer sulfur concentrations, which are a significant factor in reducing atmospheric visibility, vary significantly over a few hours of time at any site and, between sites in a network, show correspondences that suggest long range transport.

In Florida, a north-to-south average concentration decrease is observed, which may result from the averaging or mixing of polluted continental air with cleaner marine air. However, the Bermuda marine atmosphere contains substantially greater fine particle sulfate concentrations than found in more remote locations, e.g., Samoa and continental South America, indicating significant urban pollution transport over the North Atlantic, 1,000 km or more from land.

From shipboard, between New York and Bermuda, anomalously high relative concentrations of aerosol sulfur in < 0.25 μm aerodynamic diameter particles are observed in parcels of air that had passed over polluted northeastern U.S. urban areas 1-2 days previously, suggesting conversion of SO_2 to sulfate over the ocean.

Within the plume from a large coal-fired power plant in New Mexico, the rate of SO_2 conversion to sulfate can be estimated from observed increases in aerosol sulfur to fly ash element ratios with time and distance up to 23 kilometers from the stack. The rate, ~ 0.7% per hour, which is calculated assuming average plant emissions during the sampling period, is consistent with the large-scale formation and transport of sulfate in urban plumes over the eastern U.S. and out over the North Atlantic Ocean.

At nonurban eastern U.S. sites, aerosol sulfur concentrations often vary inversely with relative humidity and directly with dew point and, in addition, show diurnal patterns as well as maxima during passage of cold fronts. These variations suggest that sulfate concentrations in the urban plume can vary during transport, as a result of both chemical and dynamic processes.

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Park, N.C.): I don't recall s
when ozone was present.
P. K. MUELLER: Did yo
another plume?
T. G. ELLESTAD: For th
plume passages.
P. K. MUELLER: Do you
T. G. ELLESTAD: No, I'm
quite up to making accurat
R. L. TANNER (*Brookha*
you have any ammonium da
sulfur with some more anal
T. G. ELLESTAD: The
24-hour basis. We ran a hi
head impactor and, from
ammonium values. Howev
resolution data.
J. W. WINCHESTER (*Flo*
very nice paper and a very
For the aerosol composit
mode were correlated. We h
from those of sulfur in the p
are sure that phosphorus wa
the x-ray spectrum resoluti
T. G. ELLESTAD: I'll ha
expert on the x-ray fluoresc
J. W. WINCHESTER: I'll
might be worth scrutinizin
problem.
My second comment is
indicated, of course, that it
automobiles, but St. Louis
smelting. In our analysis of
lead in the city is due to the
might be best to say "most
T. G. ELLESTAD: That

*This is a compilation of the
section.

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